

# Flow pattern in the vicinity of self-propelling hot Janus particles

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We study the temperature field and the resulting flow pattern in the vicinity of a heated metal-capped Janus particle. If its thickness exceeds about ten nanometers, the cap forms an isotherm and the flow pattern comprises a quadrupolar term that decays with the square of the inverse distance  $\sim r^{-2}$ . For much thinner caps the velocity varies as  $\sim r^{-3}$ . These findings could be relevant for collective effects in dense suspensions and for the circular tracer motion observed recently in the vicinity of a tethered Janus particle.

PACS numbers: 82.70.Dd, 66.10.cd, 47.15.G-

*Introduction.* The design of artificial micro- and nano-swimmers that propel themselves in a viscous fluid is a key issue in nanotechnology [1]. In the realm of biology, autonomous motion of microorganisms is ubiquitous and relies on surface waves or periodic body deformations [2]. Several swimming devices inspired by living systems have been built recently [3, 4], although their actuation mechanism requires external forces or torques. An alternative way toward self-propulsion is achieved using colloidal particles with non-uniform surface properties [5]. This class of rigid swimmers relies on phoretic transport, *i.e.* the force-free motion driven by the gradient of an external field [6]. In the case of self-phoresis, however, asymmetric particles are able to generate their own gradient within an otherwise homogeneous medium and thus to convert the available energy into mechanical work [7, 8].

The simplest realization of autonomous swimmers is obtained with Janus particles, which are colloidal objects with two sides differing in their physical or chemical properties [9]. For example, a bimetallic particle in a peroxide solution generates an electrochemical gradient which in turn gives rise to a flow in the surrounding fluid and thus causes self-propulsion [10]. At short times this results in linear motion, whereas at longer times the random reorientations lead to enhanced diffusion [11, 12]. Similar findings have been reported for photophoresis of hot Janus particles, which move in their own temperature gradient, with an effective diffusion coefficient that increases linearly with the heating power [13–15].

Heating of metal capped Janus particles provides a versatile means of actuation which, in particular, can be switched on and off almost instantaneously. Heat absorption of the metal cap is achieved upon illumination by a defocused laser beam [13–16] or when subject to an alternating magnetic field [17]. The metal patch absorbs the energy and converts it into heat; asymmetric heat release then drives the colloid via a mechanism of thermophoresis [6].

In this Letter we address the temperature profile and the fluid velocity in the vicinity of a hot Janus particle. In view of its large thermal conductivity we treat the metal cap as an isotherm and, as a consequence, obtain a class of hydrodynamic multipoles that are absent when

neglecting heat conduction in the cap. These additional terms result in a flow pattern which is strongly asymmetric with respect to the particle midplane, and could affect the hydrodynamic coupling between neighboring swimmers or with a bounding wall [18–20]. On the other hand, a fixed Janus particle is expected to act as a micro-pump. Visualization of the local convective flow by particle tracking velocimetry indeed revealed vortices close to a Janus particle tethered on a glass support [13]. The description of tracers trajectories thus requires a detailed knowledge of both the temperature and the velocity fields around a Janus particle.

*Temperature field.* In a first step we derive the temperature profile from Fourier's law

$$\kappa \nabla^2 T = q(\mathbf{r}) , \quad (1)$$

where  $q$  is the power absorbed by the metal cap, and

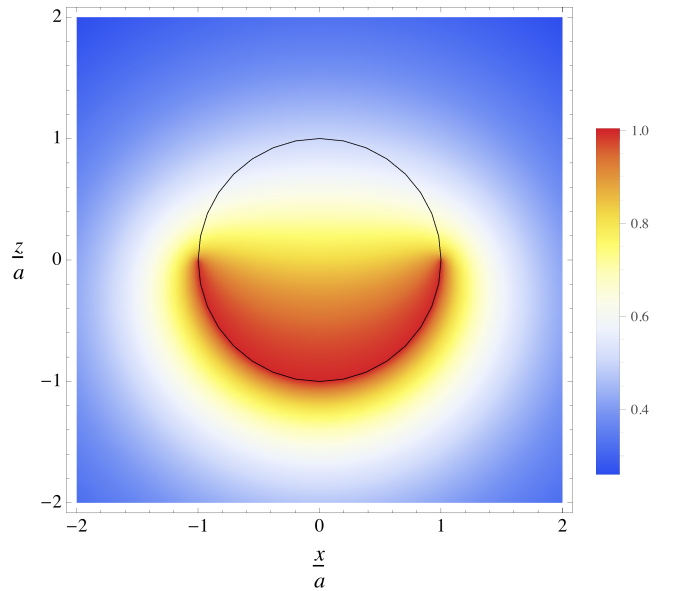


FIG. 1: (Color online) Map of the reduced temperature field  $(T - T_0) / \Delta T$  in the  $(xOz)$  plane, inside and outside the metal-coated colloid.

$\kappa$  the thermal conductivity of both the particle and the surrounding fluid (taken to be the same for simplicity.) If cap conductivity  $\kappa_c$  is usually much higher than  $\kappa$ ; if their ratio is larger than the ratio of particle radius and cap thickness,  $\kappa_c/\kappa > a/d$ , the cap forms an isotherm. Since this condition is satisfied for a 50 nm gold cap on micron-size silica or polystyrene beads, we assume in the following a constant cap temperature  $T_0 + \Delta T$ . Typical values for the excess temperature  $\Delta T$  with respect to the bulk are of the order of a few Kelvins.

Because of the mixed boundary conditions, constant temperature on the metal cap and heat flux continuity on the upper hemisphere, there is no straightforward solution of Eq. (1). As shown in the supplementary material [21], the global constraint on the isotherm can be implemented by a method based on auxiliary functions. Here we merely quote the temperature profile in the liquid phase ( $r > a$ )

$$T(r, \theta) = T_0 + \frac{\Delta T}{\pi} \sum_{n=0}^{\infty} t_n P_n(c) \left(\frac{a}{r}\right)^{n+1}, \quad (2a)$$

with  $c = \cos \theta$  and the Legendre polynomial  $P_n$ . The coefficients  $t_n$  are given by

$$t_{2k} = -t_{2k+1} = \frac{(-1)^k}{2k+1}, \quad (3)$$

except for the first one that reads  $t_0 = 1 + \pi/2$ . A similar expression with the same coefficients is found inside the particle ( $r < a$ ), albeit with  $(r/a)^n$  instead of  $(a/r)^{n+1}$ . Identifying the power  $\mathcal{P}$  absorbed by the metal cap with the total outward heat flow, one readily establishes the relation with the excess temperature:  $\mathcal{P} = (2\pi + 4)\kappa a \Delta T$ , with  $\kappa$  the thermal conductivity of the liquid. The map of the temperature field is shown in Fig. 1. The role of the isotherm assumption is illustrated by comparing with the case of a very thin cap where  $\kappa_c/\kappa < a/d$ . Then the heat conductivity of the metal structure can be neglected, and one readily finds that the even coefficients of the temperature profile vanish,  $t_{2k} = 0$  [13].

**Boundary velocity.** The temperature gradient modifies the particle-solvent interactions in a boundary layer of thickness  $\ell$ . For electric-double layer forces  $\ell$  is given by the Debye length, and for depletion forces by the gyration radius of the polymers. In both cases  $\ell$  is much smaller than the radius  $a$  of micron size colloidal particles, such that the flow pattern in the liquid can be evaluated in boundary layer approximation [6, 22, 23]. The excess enthalpy density  $h$  results in a quasi-slip velocity of the liquid with respect to the particle [24].

The boundary velocity is proportional to the temperature gradient parallel to the surface of the particle  $u_s = -(\ell^2 \bar{h} / \eta T_0) \nabla T_{||}$ , where  $\eta$  is the viscosity and  $\bar{h}$  the characteristic value of the excess enthalpy. With Eq. (2) one has

$$u_s(\theta) = u_0 \sum_{n=1}^{\infty} t_n \frac{dP_n(c)}{d\theta}, \quad (4)$$

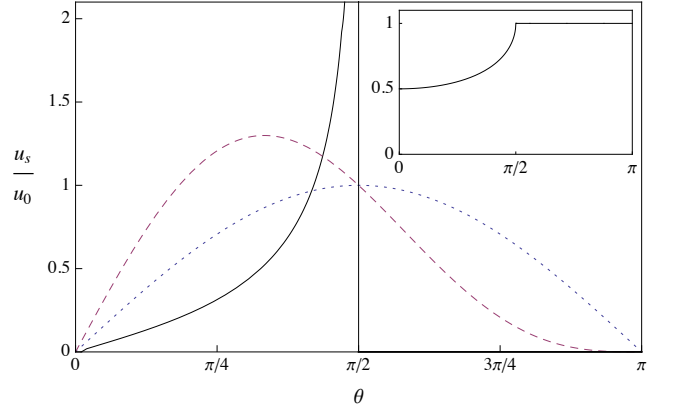


FIG. 2: (Color online) Quasi-slip velocity  $u_s(\theta)$  as a function of the polar angle  $\theta$ . For comparison, we plot the infinite series (4) (solid line), the dipolar approximation (series truncated at  $n = 1$ , dotted line) and the quadrupolar approximation (series truncated at  $n = 2$ , dashed line). Inset: surface temperature  $[T(a, \theta) - T_0]/\Delta T$  as a function of  $\theta$ .

where the prefactor  $u_0$  gives the velocity scale,

$$u_0 = -\frac{\ell^2 \bar{h}}{\pi \eta a} \frac{\Delta T}{T_0}. \quad (5)$$

The first term in Eq. (4) corresponds to the dipolar approximation:  $u_s(\theta) = u_0 \sin \theta$  [6]. Keeping the first two terms of the series, the surface velocity is that of a “squirmers” with positive stresslet  $\beta = 2/3$  [19, 25].

For positive slip velocity  $u_0$ , i.e., negative enthalpy  $\bar{h}$ , the liquid flows toward the warmer side of the Janus particle. Note that  $u_s$  is largest on the upper half-sphere close to mid-plane; it vanishes on the lower half-sphere because of the constant temperature of the metal cap – see Fig. 2. The expression  $\ell^2 \bar{h}$  has the dimension of a force, and has been evaluated for several thermophoretic mechanisms. Ruckenstein pointed out the positive slip velocity ( $u_0 > 0$ ) due to the enthalpy of the electric double layer,  $\ell^2 \bar{h} = -\frac{1}{2} \varepsilon \zeta^2$  [26], with the surface potential  $\zeta$  and the solvent permittivity  $\varepsilon$ . In many instances, however, the slip velocity is dominated by the thermoelectric effect  $\ell^2 \bar{h} = \frac{3}{2} \varepsilon \zeta S T_0$ , where the electrolyte Seebeck coefficient  $S$  may take either sign [27–29]. Upon adding polymer to the solution, thermal depletion forces result in  $u_0 < 0$  [30]. For a micro-size particle with  $\Delta T = 1$  K, the slip velocity is a few microns per second.

**Bulk velocity field.** The quasi-slip velocity on the surface of the particle induces a flow in the surrounding liquid. The general axisymmetric solution  $\mathbf{v} = v_r \mathbf{e}_r + v_\theta \mathbf{e}_\theta$  of the Stokes’ equation has been known for a long time [25, 31]. Here we give the series expansion of Ref. [32], where the radial and tangential components

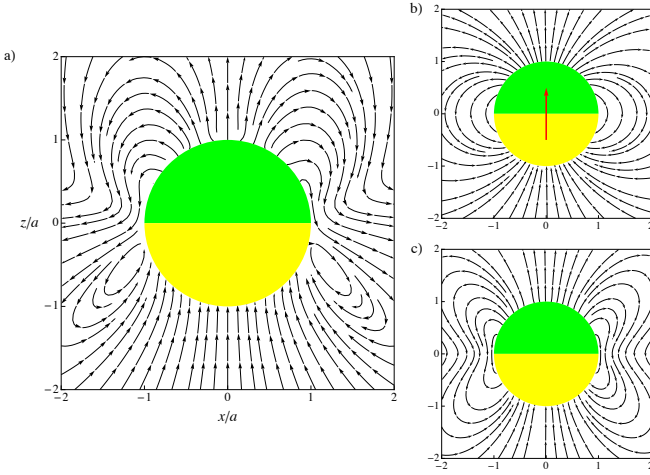


FIG. 3: (Color online) Flow streamlines around a moving Janus particle in the laboratory frame. a) The cap forms an isotherm with  $t_k$  as in (3). b) Dipolar approximation with  $t_1$  only. c) Thin-cap limit, with  $t_{2k} = 0$ .

are given by

$$v_r = u_0 \sum_{n=1}^{\infty} \frac{a^n}{r^n} \left( p_n + q_{n+2} \frac{a^2}{r^2} \right) P_n(c), \quad (6a)$$

$$v_{\theta} = u_0 s \sum_{n=1}^{\infty} \frac{a^n}{r^n} \left( p_n \frac{n-2}{n(n+1)} + \frac{q_{n+2}}{n+1} \frac{a^2}{r^2} \right) P'_n(c), \quad (6b)$$

with  $P'_n = dP_n/dc$  and  $s = \sin \theta$ . The corresponding pressure field is given in [21]. The coefficients  $p_n$  describe the inhomogeneous solutions of Stokes' equation with finite pressure, whereas the  $q_n$ 's are related to the zero-pressure homogeneous solutions. The coefficients are set by the boundary conditions at the surface of the particle. First, the far field  $\mathbf{v}$  has to match the sum of the particle velocity  $u_p \mathbf{e}_z$  and the quasi-slip velocity

$$\mathbf{v}|_{r=a} = u_p \mathbf{e}_z + u_s \mathbf{e}_{\theta}. \quad (7)$$

The second condition is a global constraint and involves the total force  $F_z = -4\pi\eta u_0 a p_1$ . In the following we evaluate the coefficients for a particle that is either freely moving or fixed at a given position.

*Moving particle.* First we consider a free Janus particle that self-propels due its own temperature gradient. Since there is no external force, the global constraint imposes the well-known condition  $p_1 = 0$  [6]. Yet, the no-force condition does not affect the inhomogeneous coefficients of higher order. Noting  $\mathbf{e}_z = c \mathbf{e}_r - s \mathbf{e}_{\theta}$ , one obtains the radial and tangential projections of Eq. (7),  $v_r = cu_p$  and  $v_{\theta} = -su_p + u_s$ . Inserting Eqs. (6a) and (6b), one readily gets for  $n = 1$

$$p_1 = 0, \text{ and } q_3 = -\frac{2}{3} t_1 = \frac{2}{3}, \quad (8)$$

and for higher orders

$$p_n = -q_{n+2} = \frac{n(n+1)}{2} t_n \quad (n \geq 2). \quad (9)$$

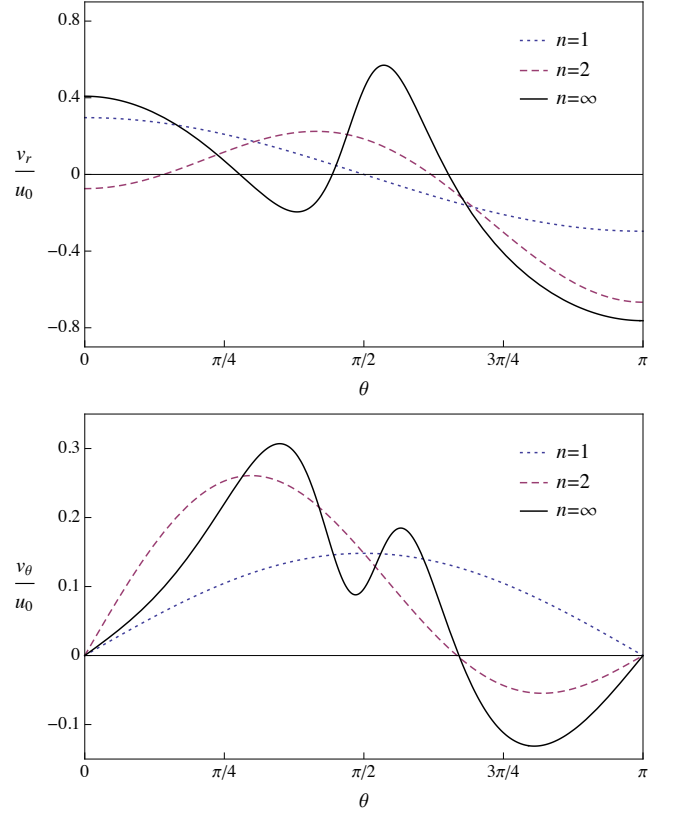


FIG. 4: (Color online) Radial and tangential components of the velocity of a moving particle at distance  $r = 1.5a$  from its center. The plots correspond to the series (6a) and (6b) truncated at  $n = 1$  (dipolar approximation, dotted lines), truncated at  $n = 2$  (quadrupolar approximation, dashed lines), and to the infinite series (solid lines).

The particle velocity is then in opposite direction to the quasi-slip and is equal to two thirds of its amplitude

$$u_p = q_3 u_0 = \frac{2}{3} u_0. \quad (10)$$

This implies that self-propulsion is driven by the dipolar term  $q_3$  only; higher Fourier coefficients of the temperature gradient,  $t_n$  with  $n > 1$ , do not contribute to the particle velocity.

We emphasize two major differences with respect to the thin-cap limit, where  $t_{2n} = 0$  and the dipolar approximation where  $t_1$  is the only non-zero coefficient. First, the temperature coefficient  $t_2$  results in a radial velocity contribution that decays with the square of the inverse distance and shows quadrupole characteristics. In the thin-cap and dipolar approximations the velocity decays as  $r^{-3}$ . Second, Fig. 3a) shows that the rotational patterns of the stream lines are located close to the metal cap; for comparison, we also plot the dipolar flow field with the only coefficient  $q_3$  and the thin-cap limit with  $t_{2k} = 0$ . The corresponding streamlines in Fig. 3b) and c) are symmetric with respect to midplane.

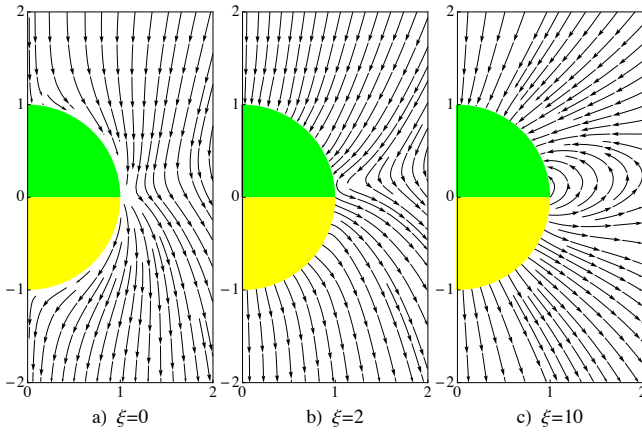


FIG. 5: (Color online) Map of the tracer velocity  $\mathbf{u}_t$  for different values of the parameter  $\xi$ .

In Fig. 4 we plot both  $v_r$  and  $v_\theta$  as a function of  $\theta$  at a distance  $r = 1.5a$  from the center of the particle; we compare the whole series with the dipolar approximation ( $n = 1$  only), and the quadrupolar approximation ( $n = 1, 2$ ). The dipolar terms are simply given by sine and cosine functions. The quadrupolar correction is by no means small or insignificant; for example,  $v_r$  changes sign at small  $\theta$ , and  $v_\theta$  at angles close to  $\pi$ . Retaining the higher-order corrections again changes the flow pattern drastically. As the most striking feature, note the large positive derivative  $dv_r/d\theta$  close to midplane; together with the positive value of the tangential component  $v_\theta$  this implies the existence of vortices at the edge of the metal cap.

*Immobile particle.* Now we turn to the situation where the Janus particle is fixed at a given position. This requires a finite external force that counteracts the self-propelling surface stress in the boundary layer. The particle velocity is then zero,  $u_p = 0$ , so that the quasislip velocity matches to the tangential component of the far-field  $u_\theta = v_\theta$ , whereas the radial component vanishes  $v_r = 0$ . One obtains the coefficients for the flow pattern

$$p_n = -q_{n+2} = \frac{n(n+1)}{2} t_n \quad (n \geq 1). \quad (11)$$

With the coefficient  $p_1 = -1$  one finds the external force  $F_z = 4\pi\eta a u_0$ . It is required to immobilize the particle which otherwise would move at a velocity  $u_p = \frac{2}{3}u_0$ , and thus corresponds to the well-known Stokes drag  $6\pi\eta a u_p$ .

In Fig. 5a) we plot the flow pattern  $\mathbf{v}(r, \theta)$ . Contrary to that of the moving particle, there are no vortices close to the particle; the liquid flows smoothly around the immobile particle. Comparison of the coefficients shows that this difference is only due to the lowest-order coefficients  $p_1 = -1$  and  $q_3 = 1$ ; in other words, the large long-

range contribution  $p_1$  hides the vortices that accompany a moving particle but are invisible in the case where the particle is fixed.

*Motion of a tracer particle.* Finally, we consider a small tracer in the neighborhood of a fixed Janus particle. Its velocity  $\mathbf{u}_t$  is given by the sum of the convective flow and of thermophoretic drift in the temperature gradient of the Janus particle

$$\mathbf{u}_t = \mathbf{v}(\mathbf{r}) - D_T \nabla T, \quad (12)$$

with  $\mathbf{v}(\mathbf{r})$  given by Eqs. (6) and (11). The mobility coefficient  $D_T$  is expressed by the enthalpy density and thickness of the boundary layer of the tracer [23]. The vector fields  $\mathbf{v}$  and  $\nabla T$  have different characteristics: at large distance, the first is isotropic and the second one of quadrupolar symmetry; close to the particle higher order terms lead to an even more intricate variation. The relative importance of the two terms in (12) is expressed by the mobility ratio of tracer and Janus particle,  $\xi = D_T/\hat{D}_T$ , which depends on their surface properties. Either term in Eq. (12) may be dominant, and they may even carry opposite sign.

In Fig. 5 we plot the tracer velocity  $\mathbf{u}_t$  for  $\xi = 0, 2$  and  $10$ . As the most striking feature, the tracer is pushed toward the colder half of the Janus particle from above but are strongly repelled from the warmer side. For  $\xi = 0$  (no thermophoresis), the tracer first flows toward the Janus particle, then creeps slowly toward the metal cap, and finally is repelled from it. For intermediate value  $\xi = 2$ , transport alongside the surface has ceased and tracer particles either accumulate at the upper side or are pushed away from the lower side. For the larger value  $\xi = 10$ , the flow pattern shows additional vortices close to the midplane of the Janus particle, so that tracers are brought back to the colder side.

Experimentally, flow circulation around a heated Janus particle tethered to a glass surface has been reported recently [13]. Tracking of fluorescent particles moreover revealed that the concentration of tracers is higher on the non coated side and lower on the coated side. It is thus likely that the observed flow pattern results from the competition between convection and thermophoresis, as expressed by Eq. (12).

In summary, we have characterized the flow around a heated Janus colloid. The discontinuity of surface properties has a major impact on the fluid velocity field not only in the vicinity of the particle but also in the bulk. In particular, we have shown that the dipolar approximation which is usually considered for simplicity is only a poor approximation of the full series. Taking into account higher order terms leads to a complex flow field that can be relevant at finite concentration, where collective effects come into play [20, 33].

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